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INHALATION EXPOSURE IN THE HOME TO VOLATILE ORGANIC CONTAMINANTS OF DRINKING WATER

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ABSTRACT

Our field studies show that indoor air concentrations of volatilized trichloroethylene (TCE) can be substantial when TCE-contaminated water is used domestically. Using a model shower, increases in TCE water concentrations, water temperature and drop path (time) increased the steady-state air TCE concentrations. Volatilization was incomplete and the rates were comparable to predicted ones. Indoor air models show that the inhalation route of exposure for such chemicals has the potential for being much greater than by direct ingestion. This should be considered in developing regulations to limit adverse health impacts from contaminants of potable water.

INTRODUCTION

There is a growing awareness of the presence of a wide range of chemicals that can occur in the home environment and cause a continuous, although normally low-level human exposure. The regulation of the chemical quality of public water supplies has been based on the assumption that the ingestion of such water constitutes the only or at least principal route by which an adverse human exposure can occur. The question arises, can potable waters brought into the home contribute significantly to these exposures by routes other than ingestion, specifically as a result of the volatilization of chemicals into the indoor air, resulting in an inhalation dose; also due to the absorption of these chemicals through the skin, primarily during bathing when a large portion of the body surface comes in contact with the water for substantial periods of time? Second, if such exposures can be expected to occur and are significant, how do they compare quantitatively to those from the direct ingestion of water?

In recent years there has been a large number of studies and assessments of indoor air pollution, such as that by a committee of the National Research Council (1981), and more recently by Spengler and Sexton (1983). These list a variety of sources of indoor air pollutants, including consumer products, materials of construction, combustion sources and tobacco smoke, but rarely refer to potable water as a possible vehicle or source, other than for radon. Concern about indoor radon exposure from water has reached the point of discussion by the U.S. Environmental Protection Agency (1983) in an advance

notice of proposed rulemaking (ANPR) concerning its possible revision of primary drinking water regulations. It is noted there that "airborne exposure from radon released into the home from water might be more significant than direct ingestion from drinking water;" also, that "it appears that radon may contribute one of the most significant cancer risks of any substance in drinking water." They indicate that maximum contaminant limits are under consideration and request comments on the need for such regulation of radon.

The U.S. Environmental Protection Agency (1982) noted similarly in an ANPR that it was considering the need to regulate volatile synthetic organic chemicals in drinking water. However, neither in the 1982 or 1983 ANPR did the discussion consider the possible inhalation exposure route in the home for these volatile chemicals, other than for radon. Similarly, dermal exposures were not discussed. Also these routes of exposure have been considered elsewhere only rarely.

The purpose of this paper is to consider the likely extent of the inhalation routes of exposure to chemicals in potable water brought into the home. Skin absorption of such chemicals during bathing has been discussed by others (Brown et al., 1984), indicating that this route of exposure may be comparable to that from the direct ingestion of water.

HUMAN EXPOSURE PARAMETERS AND AIR AND WATER USES

In order to assess human exposure to and intake of chemicals from water within a home it is necessary to have an understanding of the quantities and the ways in which such water is used. Both of these are highly variable. Table 1 shows a range of such typical daily per capita domestic uses in the U.S. These values are normally multiplied by the number of people living within a home to estimate the quantity of water brought into the home on a daily basis. Using these data for a family of four, the range is about 160 to 280 gallons or 600 to

TABLE 1
TYPICAL DAILY U.S. DOMESTIC WATER USE PER CAPITA (BOND ET AL., 1973)

Use	Quantity per capita per day	
	Gallons	Liters
Drinking and cooking	1-2	4-8
Dishwashing	1-4	4-15
Garbage disposal unit	0-4	0-15
Laundering, cleaning	3-9	11-34
Bathing	10-25	38-95
Toilet	24	91
Total	39-68	148-257

1000 liters per day. It is apparent from Table 1 that the quantities of water used in the home that most directly relate to ingestion (drinking and cooking) are a very small part of the total water usage. Much larger quantities of water are used that can and do result in volatilization of chemicals in the home and their subsequent inhalation.

Just as the water uses within a home are highly variable, so are the daily quantities of water ingested by humans. The International Commission for Radiological Protection (ICRP) has assessed various reports on the ranges of such intakes measured for adults and children under a variety of conditions such as ambient air temperature and nature of human activity (ICRP, 1975). Direct intake of tap water itself, in contrast to water intake from milk or water-based drinks (tea, coffee, bottled beverages), can be small. The ICRP reports that under normal conditions the direct intake of tap water ranges from 45 to 730 mL per day for adults, compared to a total fluid (water) intake range of 1000 to 2400 mL. More recently the variability of drinking water intakes by adults from studies in Canada, Great Britain, the Netherlands and New Zealand has been reported (Gillies and Paulin, 1983). Among these studies the mean daily intakes, including those for beverages made with tap water, ranged from 0.96 to 1.30 liters per day. In one of the New Zealand studies the relative standard deviation of such intakes for about 2000 adults was 570 mL per day, the mean being 960 mL per day. In order to have a standardized basis for assessing the quantities of contaminants in potable water that may be ingested, the ICRP has developed a fluid (water) intake table for reference man (Table 2). For reference adult man, adult woman and child the tap water reference intake is in each case less than 15 percent of the total fluid (water) intake.

TABLE 2
DAILY FLUID INTAKE FOR REFERENCE MAN (ICRP, 1975)

Source	Fluid intake (mL per capita per day)		
	Adult man	Adult woman	Child (10 yr)
Milk	300	200	450
Tap water	150	100	200
Other	1,500	1,100	750
Total fluid	1,950	1,400	1,400

The 1950 mL per capita total fluid intake for reference adult man is approximately equal to the 2-liter quantity frequently used to establish standards for and assess health effects from exposures to chemicals and other constituents in public water supplies. It is conservative to do so (in a

protective sense). Nevertheless, to assess more accurately such impacts, particularly in relation to other human inhalation exposures to chemicals in water, the types of waters ingested and their variabilities should be considered.

Although respiratory rates can also be quite variable, the ICRP has developed a set of rates and volumes for reference man air intake, as shown in Table 3. Although the 10 m^3 per day value corresponding to the respiratory volume for adult man of 9.6 m^3 in an 8-hour working period is often used in assessing daily exposures in the workplace, for air exposures within a home a different value should be used normally, depending on the fraction of the day spent in the home and the activities undertaken. Thus, for example, an adult not leaving the home might appropriately be considered to have a daily respiratory volume of about 20 m^3 , twice the amount normally used for an 8-hour work exposure period. At the same time, where there is a shorter period of unusual air exposure, such as in a shower or bathroom contaminated with a volatile chemical, a short-term respiratory rate is of interest. From Table 3, a value of 1 to 1.2 m^3 per hour is a good estimate for adult reference man or woman in this regard.

TABLE 3
RESPIRATORY VOLUMES FOR REFERENCE MAN, CUBIC METERS OF AIR BREATHED (ICRP, 1975)

Activity	Respiratory volume (m^3)			
	Adult man	Adult woman	Child (10 yr)	Infant (1 yr)
8-hr working light activity	9.6	9.1	6.2	2.5 (10 hr)
8-hr non-occupational activity	9.6	9.1	6.2	-
8-hr resting	3.6	2.9	2.3	1.3 (14 hr)
Total (m^3/day)	23	21	15	3.8

Such estimates for water and air exposures do not address the question of the absorption of chemicals from the air and water media taken in via ingestion or inhalation. The relative absorption factors for a given intake by these two routes of exposure can be quite variable and specific to each chemical. These will not be addressed. Our principal focus will be exposure, which we define as the quantity of a chemical contained in a given amount of inhaled air or ingested water.

PHYSICO-CHEMICAL ASPECTS

The volatilization of chemicals from water used within buildings has both equilibrium and time (rate) components that should be considered in estimating the indoor air concentrations that can ensue. These in turn will interact with the air movement factors, such as the rates of forced air ventilation, infiltration and exfiltration to establish the air concentrations to which people will be exposed.

The equilibrium and kinetic aspects of such volatilization have been assessed for a variety of low solubility chemicals that may be encountered as contaminants of natural and treated fresh waters (MacKay and Wolkoff, 1973). They showed that a convenient expression for the equilibrium of the volatile chemical between the air and water phases is

$$P_i = C_i P_{is} / C_{is} \quad (1)$$

where P_i is the air concentration (atm) of the compound "i" in equilibrium with its water concentration C_i (mol/m³, essentially millimol/L); P_{is} is the vapor pressure of the pure compound and C_{is} its solubility in water. The ratio P_{is}/C_{is} is the well known Henry's law constant H_i . Thus

$$H_i = P_{is} / C_{is} \quad (2)$$

and therefore

$$P_i = H_i C_i \quad (3)$$

This is Henry's law which states simply that the equilibrium vapor pressure of a volatile constituent above an aqueous solution is directly proportional to its concentration in that aqueous solution. Although the Henry's law constant has been measured for some anthropogenic chemicals, Mackay and Wolkoff's analysis is useful in that it is a tool to predict values of H_i for chemicals of limited water solubility from other measured quantities, namely the vapor pressure of the pure chemical and its solubility in water.

Temperature usually affects equilibrium constants such as H_i . The vapor pressure of pure compounds will increase with temperature. Solubilities of chemicals in water can either decrease or increase with temperature. Over the temperature range of 20 to 30 °C carbon tetrachloride solubility in water is relatively constant at about 800 mg/L, while chloroform decreases in solubility slightly with increased temperature from about 8.1 to 7.6 g/L over the same temperature range (National Academy of Sciences, 1978). Yet over that temperature range the vapor pressures of the pure liquids increase substantially

with temperature, from 90 to 140 mm mercury for carbon tetrachloride, and from 155 to 246 mm mercury for chloroform. Thus Equation 2 indicates that for each of these chemicals the net effect is that H_i will increase over this temperature range; and for a given concentration in water the equilibrium vapor pressure for each of these constituents will similarly increase. This effect can be important in establishing the extent to which such chemicals in water brought into a home may volatilize and be affected by the temperature of the water use.

Mackay and Wolkoff (1973) further discuss how the rate of evaporation of such chemicals from dilute aqueous solution can be expressed by a first order differential equation:

$$dC_i/dt = kM_iH_iC_i \quad (4)$$

where k is a proportionality constant including such factors as the rate of evaporation of water and its vapor pressure; M_i is the molecular weight of the evaporating chemical; and t is time. When the volatilizing chemical in the air above the water in a home is below the equilibrium vapor pressure, there is a "driving force" to volatilize. As the air concentration of the chemical builds up and approaches equilibrium, the reverse process becomes important, so that the net evaporation rate decreases.

These concepts have been utilized in interpreting the results of experimental studies of the transfer of six organic chemicals (listed in Table 4) from aqueous solution to the atmosphere, using oxygen as a reference compound (Roberts and Dandliker, 1983). The chemicals listed there are in order, dichloro-difluoromethane, carbon tetrachloride, tetrachloroethylene, 1,1,1-trichloroethane, trichloroethylene (TCE) and chloroform, most of which are of

TABLE 4
AIR-WATER EQUILIBRIUM CONSTANTS AND TRANSFER RATE COEFFICIENTS FOR
SEVERAL VOLATILE ORGANIC CONTAMINANTS (ROBERTS AND DANDLIKER, 1983)

Compound	Henry's const. (atm x m ³ /mol)	Mass trans. prop. coeff. ^a
CCl ₂ F ₂	1.5	0.66
CCl ₄	2.4 x 10 ⁻²	0.62
CCl ₂ =CCl ₂	1.5 x 10 ⁻²	0.61
CH ₃ CCl ₃	1.5 x 10 ⁻²	0.61
CHCl=CCl ₂	9.9 x 10 ⁻³	0.62
CHCl ₃	5.3 x 10 ⁻³	0.56

^aCompared to oxygen; see text

interest in that they have been found to be present in many groundwaters and public water supplies.

As shown in Table 4, aside from the first compound, the Henry's law constants, H_i , do not vary by more than a factor of about five. However, a more varied group of compounds of limited water solubility can have a wide range of values of H_i (Mackay and Leinonen, 1975). For example, they reported values of 2×10^{-7} , 4×10^{-5} , 6×10^{-4} , and 7×10^{-3} for dieldrin, DDT, Aroclor 1242 (a PCB) and toluene, respectively, at 25 °C. Thus in order to assess a possible equilibrium situation that might be encountered, such as in a home, the H_i value for the specific compound must be utilized, rather than assuming that it might lie in the relatively narrow range of values listed in Table 4.

Roberts and Dandliker (1983) evaluated their evaporation rate experiments in terms of a traditional mass-transfer rate expression:

$$dC_i/dt = K_L A C_i \quad (5)$$

where K_L is the overall mass-transfer rate coefficient and A is the specific surface area (m^2) per volume of aqueous solution from which the solute is evaporating. This is similar to Equation 4, but states explicitly that the rate of evaporation will necessarily increase in proportion to the surface area of the solution. Roberts and Dandliker note that the mass transfer rate coefficient K_L may be influenced by both the liquid- and gas-phase resistances (to diffusion) at the air-water interface. Following the approach of Mackay and Leinonen (1975), they estimated that for all compounds with H_i greater than 4.8×10^{-3} , which includes all of those listed in Table 4, liquid phase resistance is rate limiting. Interpreting their experimental results in terms of Equation 5, they defined a mass transfer proportionality constant essentially as the ratio of K_L for each compound divided by that for oxygen. In Table 4 it is seen that for each compound this has a value of about 0.6, consistent with the fact that for each of these compounds the diffusivity in water is about 0.6 times that of oxygen (Roberts and Dandliker, 1983), and therefore that the rate controlling step in these experiments is liquid phase diffusion. This then provides the basis for relating mass transfer rates from water surfaces to the diffusion coefficients (diffusivity) in the aqueous phase. However, the authors note that additional work is needed to assess the possible influence of gas-phase resistance in practical applications.

INDOOR AIR MODELING TO ESTIMATE RELATIVE AIR AND WATER EXPOSURES

Various models have been used to estimate expected indoor pollutant concentrations, including one- and multi-compartment models, as well as

empirical models based on statistical evaluation of concurrent indoor and outdoor air concentrations and other relevant terms (Wadden and Scheff, 1983). The single compartment model has been used widely in analyzing indoor air concentrations, as described by Wadden and Scheff. Two air concentration terms are used, C_i and C_o , the indoor and outdoor values, respectively. The source term S is the quantity of pollutant generated indoors per unit time (e.g., milligrams per hour). The model can encompass a pollutant decay factor R , often treated as a first-order rate proportional to C_i , and a mixing factor k , which reflects a deviation from ideal mixing within the home ($k = 1$) and which has been found to range from 0.1 to 0.33. This mixing factor is important in that, to the extent that there is "dead space" within a home, the effective indoor air volume in this model will decrease and the steady-state indoor air concentration from a pollutant source will increase.

The outdoor air concentration C_o is of importance in the model in that it will manifest itself as an indoor air concentration from makeup air and infiltration. Also, because of loss to the outdoors by exfiltration and exhaust, even if there is no decay mechanism for the pollutant or removal by filtration in air recirculation, there will be a finite buildup with time for a continuously generated pollutant.

This single compartment model in simplified form has been used to estimate the steady-state indoor air concentration of a continuously generated pollutant (Wadden and Scheff, 1983). The parameters are as follows: V , home air volume, 450 m^3 ; q_1 , indoor air recirculation rate, $1350 \text{ m}^3/\text{hr}$ (this is equivalent to three room air changes per hour); q_2 , infiltration rate, $338 \text{ m}^2/\text{hr}$ (this was taken also to be the exfiltration rate, q_3); and k , the mixing factor, 0.15. They further assumed that the outdoor air concentration of the pollutant C_o was zero and that there was no removal of gaseous pollutants by the filters. For certain chemicals, such as formaldehyde and carbon monoxide, they assumed that there were no decay mechanisms within the home.

For this set of assumptions the steady-state form of the equation based upon the model takes the simple form:

$$C_i = S/(kq_2) \quad (6)$$

Thus the steady-state indoor air concentration is directly proportional to the source term S and inversely to the mixing factor and infiltration rate. Using the values cited above for the latter terms

$$C_i = 0.02S \quad (7)$$

where C_i has the units of mass/m^3 and the source term S , mass/hr .

It should be noted that the air recirculation rate q_1 does not appear in Equation 6, nor does the indoor air volume, because it was assumed that there were no removal nor decay mechanisms within the home. When these occur, the final form of the equation becomes more complex.

Such an approach was used to relate measurements of radon within buildings to the uses of water from which the radon volatilizes (Prichard and Gesell, 1981). They utilized a simple single compartment model which they expressed in the form

$$C_i = (C_w/24q_2) \sum_j e_j W_j \quad (8)$$

where q_2 is the volume of outdoor air passed through the dwelling per hour; "j" refers to each different use of water in the home that can result in volatilization of radon; e_j is the transfer efficiency (fraction of radon present that volatilizes) in that use; W_j is the average daily quantity of water in such use; C_w is the concentration of radon in the water brought into the home; and C_i is the calculated indoor air radon concentration. The symbols for the terms in the Equation 8 have been modified to be consistent with those used in this paper. Equations 6 and 8 are essentially the same in form in that $C_w e_j W_j$ is equivalent to the source term S of Equation 6 and $24q_2$ is the daily air flow through the home, equivalent to kq_2 of Equation 6. The Prichard and Gesell model does not, however, utilize a mixing factor term k .

Equation 8 predicts that the steady-state indoor air radon concentration from water will be proportional to its concentration in the water when the other parameters are constant. Table 5 shows the result of their calculation of the

TABLE 5
RADON RELEASE FROM DOMESTIC USE OF WATER CONTAINING 1000 pCi RADON
PER LITER (PRICHARD AND GESELL, 1981)

Use	Daily consumption (liters)	Transfer efficiency (%)	Radon liberated (pCi)
Showers	150	63	94,500
Tub baths	150	47	70,500
Toilet	365	30	109,500
Laundry	130	90	117,000
Dishwasher	55	90	49,500
Drinking and kitchen	30	30	9,000
Cleaning	10	90	9,000
Total	890	-	459,000

quantities of radon released (from water containing 1000 pCi radon/L) for each water use estimated for a typical family of four, using transfer efficiencies determined experimentally. The transfer efficiencies shown there vary from 30 to 90 percent, but the average efficiency weighted by water use is about 50 percent; that is, 50 percent of all radon brought into the home in these water uses is volatilized within the home, the remainder presumably leaving in the domestic wastewater. They note that since radon is not chemically reactive, the removal patterns by air flow and otherwise follows the same pattern as the loss of any other relatively non-reactive pollutant.

They also estimated the variability in steady-state concentrations in air that could be encountered, depending on the type of building and air exchange rates. These estimates indicate that the air radon concentrations from water can be expected to vary considerably, the extremes of the range differing by a factor of 50. Finally they note that the exposure models were corroborated by a series of measurements of radon in air, the results supporting the models within experimental error.

The likely extent of volatilization or transfer efficiency in indoor water uses will be affected by the Henry's Law constant. For example, using for chloroform the H value of 5.3×10^{-3} shown in Table 4, one can calculate at 25 °C using Equation 3 and the ideal gas law that at equilibrium

$$C_A = 0.2C_W \quad (9)$$

where C_A and C_W are the respective concentrations in air and water in units of mass per volume, such as mg/L. For a typical U.S. domestic water use of 30 L/hr and an air flow rate of 3×10^5 L/hr, the ratio of air to water use V_A/V_W is 10^4 . At Henry's Law equilibrium, in such uses one would predict that the relative masses of a volatile constituent that would be distributed between the air and water, as expressed by M_A/M_W would be

$$M_A/M_W = (C_A/C_W)(V_A/V_W) \quad (10)$$

For chloroform, using Equation 9 in conjunction with Equation 10 and the value of 10^4 for V_A/V_W , one calculates a value for M_A/M_W equal to 2000. This indicates that the equilibrium for chloroform is displaced in the direction of essentially complete volatilization, as it would be for many volatile constituents. In such cases the principal factors that would affect the achievement of equilibrium are the time of water exposure to air in the home and the area of the water-air interface. The latter in turn would be influenced substantially by the hydrodynamic regime, such as the geometry and turbulence in a clothes washer.

The simple indoor air model that leads to Equations 6 and 7 can be used to generate an estimate of the maximum indoor air concentrations that are likely to result from the volatilization of chemicals from water brought into a home. For example, one can use the data from Table 1 which indicate that the per capita total daily indoor use of water is in the range of 39 to 68 gallons per day or 592 to 1028 liters per day for a family of four. Taking a value of 720 liters per day, this is equivalent to 30 liters per hour. If one assumes further that all of the volatile constituent at concentration C_w (mass per liter) in the water volatilizes into the indoor air (100 percent transfer efficiency), the source term S becomes

$$S = 30C_w \quad (11)$$

which when combined with Equation 7 leads to

$$C_i = 0.6C_w \quad (12)$$

recognizing that the concentration units are different (C_i , mass per m^3 in air; C_w , mass per L in water).

The next question is that of the relative exposures to a volatile pollutant from the ingestion and inhalation routes. Using the data from Tables 2 and 3 for reference man, and taking adult man as an example, one can use a daily respiratory volume of $20 m^3$ and a daily water intake of either 2 liters (total fluid) or 0.15 liter (tap water). The daily exposure from water to a volatile constituent is then calculated simply by multiplying the concentration in water C_w (mg/L) by either 2 or 0.15. The daily exposure from air is the product of 20 times C_i , using Equation 12 to determine the latter. These are presented in Table 6 for the water concentration of a constituent, C_w , and a specific example

TABLE 6
ESTIMATED INDOOR INHALATION AND INGESTION EXPOSURES FOR AN ADULT MALE FROM
VOLATILE CHEMICALS IN WATER USING A ONE-COMPARTMENT INDOOR AIR QUALITY MODEL

Water conc. (mg/L)	Exposure, mg/day		
	Air	Water	
		2 L intake	0.15 L intake
C_w	$12 C_w$	$2 C_w$	$0.15 C_w$
0.01	0.12	0.02	0.0015

of the latter, namely 0.01 mg/L. It is apparent from Table 6 that exposure from the air route is substantially greater than that from water ingestion using this single compartment air model and the assumptions stated above. If a daily ingestion of two liters is assumed, the air exposure is higher than that from water by a factor of 6. This ratio becomes much larger, namely 80, if the smaller estimate of 150 mL per day is used for water ingestion. One should be cautious, however, in using these ratios as more than an example of the possible relative exposures that can occur via the drinking water and air inhalation routes. Nevertheless, they do indicate the possibility of substantially greater air exposures to volatilized constituents from water used within the home and, therefore, the need to consider this route of exposure in assessing health effects from such contaminants in potable water supplies.

TRICHLOROETHYLENE IN FIELD AND LABORATORY STUDIES

We investigated the possible volatilization of trichloroethylene (TCE) into indoor air within buildings in a small community using individual wells obtaining water from an aquifer measured to contain about 40 mg TCE/L (Andelman et al., 1985). Using a continuous real-time monitor with an infra-red detector, measurements were taken in closed rooms in two homes and a small municipal building on one day in July 1983. Prior to turning on water in bathrooms, no TCE could be detected in the indoor air above the detection limit for the instrument, namely 0.5 mg/m^3 . However, TCE was readily detected in the bathrooms with water running. As expected, the air concentration levels increased with time as shown in Table 7. In home B, the highest air concentration of TCE measured after 17 minutes of the shower running was 81 mg/m^3 , approximately one-third of the eight-hour time-weighted threshold limit value of 270 mg/m^3 for the work environment (American Conference of Governmental Industrial Hygienists, 1984). To estimate the possible inhalation exposures within these homes from these shower uses, one can take a value of about 40 mg/m^3 as the average air concentration during the shower period. Assuming one-hour per week in the shower and an air breathing volume of 1.2 m^3 in that hour, this would correspond to a dose of 48 mg TCE per week (neglecting any question of the fraction of the dose that is absorbed). If one also assumes an ingestion of the contaminated water at 150 mL/day (1.05 L/week), the water dose is 42 mg TCE per week, indicating that the shower air and ingestion doses may be reasonably comparable. One would also expect an increased air dose from exposure elsewhere in the home.

To investigate further the factors that influence TCE air concentrations resulting from showers, a scaled-down model shower was constructed and operated with known concentrations of TCE injected continuously into the inlet water, typically in the range of 1.5-2.9 mg TCE/L. The experimental details and

TABLE 7
INDOOR AIR CONCENTRATIONS OF TRICHLOROETHYLENE (TCE) IN BUILDINGS USING WELL
WATER CONTAINING APPROXIMATELY 40 mg TCE PER LITER (ANDELMAN ET AL., 1985)

Location	Time	Conc., mg/m ³
Municipal Bldg. Ladies rest room, water running	12:20	35
Home A Kitchen	1:34	ND ^a
Bathroom, shower on	1:53	32
	1:56	48
	2:01	67
	2:03	72
Home B Bathroom upstairs, shower on	2:20	ND
	2:27	19
	2:30	35
	2:33	40
	2:37	81
Bathroom downstairs, shower on	3:08	ND
	3:17	64
	3:24	67

^aND - not detected above detection limit of 0.5 mg/m³

complete results are reported elsewhere (Andelman et al., 1985). The shower chamber volume was 0.1 m³ and the regulated air throughput 0.005 m³/min, equivalent to three air changes per hour. Based on the diameter of the shower head orifices, it was estimated that a spherical droplet emitted from the shower head would have a diameter of approximately 0.25 mm. The shower water flow was 0.3 L/min at a temperature which was varied between 23 and 41 °C, the drop path being adjusted from 25 to 75 cm.

Continuous monitoring of air concentrations of TCE in the shower chamber was performed by pumping air from it to either an infra-red or flame ionization detection system. Steady-state air concentrations were achieved typically within 60 minutes, after which TCE injection was discontinued. The decay in TCE air concentration was also monitored.

Increases in TCE inlet water concentration, water temperature and drop path height all increased the steady-state air TCE concentrations, as expected. Figs. 1 and 2 are typical buildup and decay curves showing the effects of the two latter parameters. In all experiments the measured air TCE concentrations were considerably lower than those predicted by Henry's Law for the corresponding water TCE concentrations. At 25 °C one can calculate such an

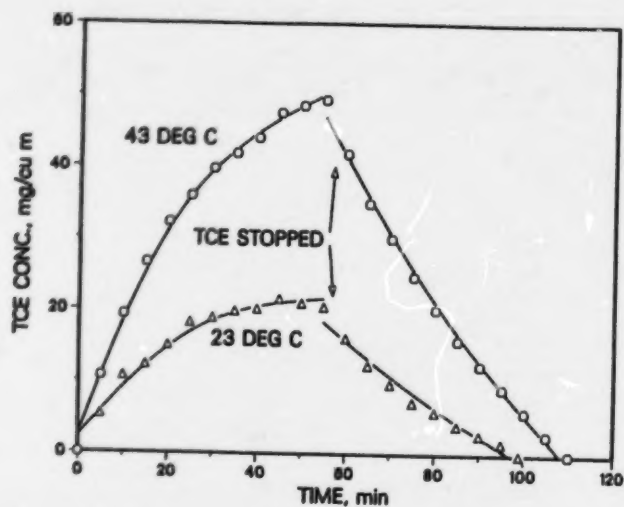


Fig. 1. Effect of water temperature on TCE air concentration in model shower.

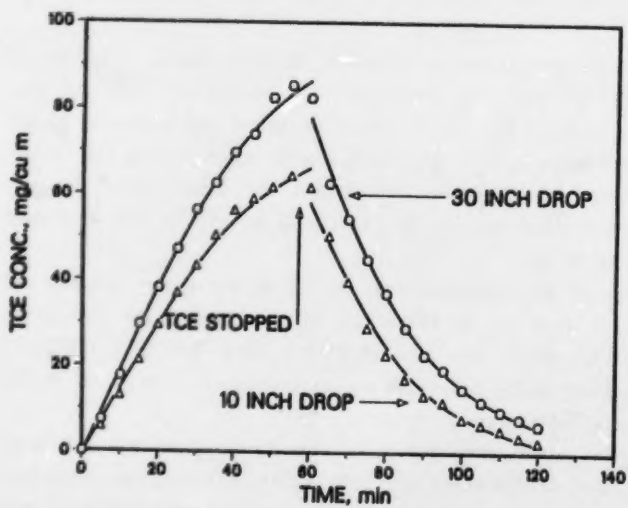


Fig. 2. Effect of drop path height on TCE air concentration in model shower.

equilibrium relationship for TCE comparable to that for chloroform in Equation 9. For TCE at this temperature C_A/C_W would thus equal 0.4. For the typical shower water concentration of 2 mg TCE/L used in these experiments, the equilibrium air concentration expected at 25 °C is then 0.8 mg TCE/L or 800 mg/m³, well above the approximately 20 mg/m³ shown in Fig. 1 for the 23 °C curve. Thus one can conclude that the rate of volatilization is sufficiently slow so that Henry's Law equilibrium is not attained and that the interplay between the kinetics of volatilization and the transport of TCE from the chamber by air passed through it establishes the TCE air concentration at a given time.

Measurements of TCE in the inlet and effluent shower chamber water indicate that typically 40-60 percent of the TCE volatilized, depending on experimental conditions. This is generally consistent with theoretical expectations, based on a model of gas transfer across an air-water interface (Liss and Slater, 1974). Assuming a probable 1.0 m/sec droplet velocity for the likely droplet size in our system, their model predicts about 20-45 percent volatilization for the 0.25-0.75 second drop time expected for the 10 inch (25 cm) and 30 inch (75 cm) droplet paths shown in Fig. 2. For many volatile solutes the rate-limiting step in the volatilization process is diffusion across the liquid film at the water-air interface. The substantial increase with temperature in the rate of TCE volatilization shown in Fig. 1 is consistent with the well known effect of temperature on diffusion of solutes in aqueous solution, including across such a liquid film.

Based on the fact that the measured air concentrations of TCE were well below those expected at equilibrium, and that 40-60 percent of the TCE volatilized from the water, one can write a mass-balance equation using a constant rate of input, K , for TCE volatilizing from the shower water per unit time. This K in fact represents a complex process of a first-order rate of volatilization which gradually decreases for a given drop of water as it moves from the shower head and reaches the bottom of the shower chamber. Nevertheless, within that short time (less than a second) one can reasonably treat the volatilization rate on an integrated average basis. The mass-balance equation is then

$$V(dC_A/dt) = K - FC_A \quad (13)$$

where V is the shower chamber volume, C_A the air concentration of TCE, and F the rate of flow of air through the chamber. This equation assumes that there is rapid and complete mixing within the chamber. (Although we have experimental evidence to the contrary, we have not yet quantified the incompleteness of mixing.) As the TCE air concentration increases, eventually the rate of removal will equal the rate of volatilization and a steady-state will be attained. At that point dC_A/dt will be equal to zero and, from Equation 13,

$$C_A(\text{steady-state}) = K/F \quad (14)$$

The function describing the build up to steady-state is obtained by integrating Equation 13 to yield

$$\ln(1 - FC_A/K) = -(F/V)t \quad (15)$$

Equation 15 describes the build-up portion of the experimental curves of Figs. 1 and 2 to the point where the TCE injection is stopped. In several of our experiments, such as the 23 °C curve of Fig. 1, it appears that steady-state was reached by the time the TCE injection was stopped. The function for the time required to attain a given fraction f of the steady-state concentration is obtained by combining Equations 14 and 15 to give

$$\ln(1 - f) = -(F/V)t_f \quad (16)$$

Taking the shower system parameters of F and V equal to 0.005 m³/min and 0.1 m³, respectively, and using Equation 16, $t_{0.9}$ has a value of 46 minutes, somewhat smaller than the approximately 55 minute time when the TCE injection was normally discontinued, and consistent with a leveling off of the TCE air concentration around that time.

The decay portion of the curve after discontinuing TCE injection is represented simply by Equation 13 with K equal to zero, the integrated form being

$$\ln(C_0/C_t) = (F/V)t \quad (17)$$

Although not shown here, the decay curves did follow this relationship with constant values for the slopes when the F and V parameters were not changed.

Extending these results from the scaled-down model shower system to full-size shower systems with their considerably longer droplet paths, one can expect that a large fraction of TCE and other highly volatile contaminants of potable water would volatilize in shower usage. This can result in an inhalation exposure, both to the user of the shower and to other inhabitants of the home as the shower air is distributed through it.

SUMMARY AND CONCLUSIONS

Air and water measurements in homes have shown the presence of trichloroethylene (TCE) and other volatile chemicals, such as radon, due to contamination of groundwater systems. Rate and equilibria relationships for such constituents volatilizing into air from water have been studied. These can

be used to estimate the extent to which this process occurs indoors.

Indoor air quality models have been developed and used to predict the steady-state air concentrations of a variety of volatile constituents originating from materials used in the home. These can be utilized for volatile chemicals and radon-222 in potable water. Using such indoor air models, calculations of inhalation exposures indicate that this route can be substantial in comparison with direct ingestion of the contaminated water.

In a model shower system using water containing TCE, the air in the vicinity of the shower water was found to increase in TCE concentration with time. The air concentrations of TCE were dependent as expected on several factors, including water temperature and height of the drop path. These experiments indicate that the resulting human inhalation exposures can be substantial for highly contaminated waters, and this has been shown in field measurements as well. Such exposures should be considered in the development of regulations to limit the possible adverse health impacts from the use of contaminated potable water.

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